What is claimed is:

 A novel crystalline ion-association substance having a general formula (I):

 $[\{C_5(R^1)_n\}_{2m}M_m]^{1+}[\{B(R^2)_4\}^{-}]_1$

wherein M is a transition metal of center nucleus; C_5 is cyclopentadienyl group; R^1 is electron donative or electron attractive substituent bonded to a carbon atom of cyclopentadienyl group, or an organic group bridging two neighboring carbon atoms; \underline{n} is a number within range of 0 to 3; \underline{m} is either 1 or 2; $\underline{1}$ is either 1 or 2; R^2 is a ligand coordinated to boron atom (B), and the four R^2 (s) are the same to each other.

- 2. The crystalline ion-association substance claimed in the claim 1 wherein said transition metal of center nucleus (M) of said general formula (I) is selected from a group consisting of Ti, Zr, Fe, Ru, Os, Hf, V, Cr, Mo and W.
- 3. The crystalline ion-association substance claimed in the claim 1, wherein said electron donative or electron attractive substituent of said general formula (I) is, identical to or different from each other, selected from a group consisting of alkyl group, cycloalkyl group, alkoxy group, aryl group, dialkyl group, silyl group, acyl group, cycloalkenyl group, amino group, carboxyl group, organoboranyl group, phosphino group, aldehyde group, hydroxyl group and vinyl group; and said organic group bridging two neighboring carbon atoms is alkylene group.
- 4. The crystalline ion-association substance claimed in the claim 1, wherein the metallocene derivative cation having mono-nucleus structure or di-nucleus structure which constitutes the crystalline ion-association substance having the general formula (I) is selected from a group consisting of acetyl ferrocenium, tert.-amyl ferrocenium, benzoyl ferrocenium, n-butyl ferrocenium, cyclohexenyl ferrocenium, cyclopentenyl ferrocenium, 1,1'-diacetyl ferrocenium, 1,1'-di-n-butyl ferrocenium, N,N-dimethylaminomethyl ferrocenium, 1,1'-dimethyl ferrocenium, ethyl ferrocenium, (dihydroxyboranyl) ferrocenium, 1-hydroxyethyl ferrocenium, hydroxymethyl ferrocenium, vinyl ferrocenium, 1,1-bis(diphenylphosphino) ferrocenium, ferrocenium, t-butyl ferrocenium, dibutyl ferrocenium, bis(cyclopentadienyl) chromium cation, bis(cyclopentadienyl) molybdenum chloride cation,

bis(cyclopentadienyl) osmium cation, bis(t-butylcyclopentadienyl) titanium chloride cation, bis(cyclopentadienyl) dicarbonyl titanium cation, bis(cyclopentadienyl) titanium chloride cation, bis(cyclopentadienyl) tungsten chloride cation, bis(i-propylcyclopentadienyl) tungsten chloride cation, vanadocenium, bis(n-butylcyclopentadienyl) zirconium chloride cation, bis(t-butylcyclopentadienyl) zirconium chloride cation, bis(cyclopentadienyl) zirconium chloride cation, bis(ethylcyclopentadienyl) zirconium chloride cation, bis(ethylcyclopentadienyl) zirconium chloride cation, bis(methylcyclopentadienyl) zirconium chloride cation, bis(indenyl) dimethyl zirconium cation, bis(t-butylcyclopentadienyl) hafnium chloride cation, bis(iso-propylcyclopentadienyl) hafnium chloride cation and diferrocenium derivative cation.

- 5. The crystalline ion-association substance claimed in the claim 1, wherein said ligand (R²) of the said formula (I) is selected from a group consisting of aryl group, halogenated aryl group, halogen haloform aryl group, cyclocalkynyl group, halogenated cycloalykl group, halogenated cycloalkynyl group, cycloalkyloxy group, cycloalkenyloxy group, alkadienyl group, alkatrienyl group, halogenated alkatrienyl group, halogenated alkatrienyl group, halogenated alkatrienyl group, halogenated alkynyl group and heterocyclic group.
- 6. The crystalline ion-association substance claimed in the claim 1, wherein said tetranentate borate complex anion which constitutes the crystalline ion-association substance having the general formula (I) is selected from a group consisting of tetrakis(4-fluorophenyl) borate anion, tetrakis(4-fluorobiphenyl) borate anion, tetrakis(3,5-bis(trifluoromethyl)phenyl] borate anion, tetrakis(3,5-difluorophenyl) borate anion, tetrakis(4-(trifluoromethyl)phenyl] borate anion, tetrakis(2,3,5,6-tetrafluorophenyl) borate anion, tetrakis(1,2,3,4,5-pentafluorophenyl) borate anion, tetrakis(3,4,5-trifluorophenyl) borate anion, tetrakis(3-fluoropropane) borate anion,

tetrakis[3,5-bis(1,1,1,3,3,3-hexafluoro-2-methoxy-2-propyl)phenyl]

borate anion, tetrakis(2,4,6-trifluorophenyl) borate anion, tetrakis(nonafluorobutyl) borate anion, tetrakis(perfluorohexyl) borate anion, tetrakis(perfluoropentyl) borate anion, tetrakis(perfluorooctyl) borate anion, tetrakis (perfluoro-3-methylbutyl) borate anion, tetrakis(perfluoro-5-methylbutyl) borate anion, tetrakis(heptafluoropropyl) borate anion, tetrakis(3,5-dichlorophenyl) borate anion, tetrakis(4-chlorophenyl) borate anion, tetrakis(benzyl chloride) borate anion, tetrakis(chlorobenzyl) borate anion, tetrakis[2-(perfluorobutyl)ethyl] borate anion, tetrakis[2-(perfluorohexyl)ethyl] borate anion, tetrakis[2-(perfluorooctyl)ethyl] borate anion, tetrakis[2-(perfluoro-7-methyloctyl)ethyl] borate anion, tetrakis[2-(perfluoro-5-methylhexyl)ethyl] borate anion, tetrakis(2,2,3,3-tetrafluoropropyl) borate anion, tetrakis(1H,1H,5H-octafluoropentyl) borate anion, tetrakis(1H-perfluorohexyl) borate anion, tetrakis(1,1-difluoroethyl) borate anion, tetrakis[3,5-bis(trifluoromethyl)benzyl] borate anion, tetrakis[4-(trifluoromethyl)benzyl] borate anion, tetrakis(3,5,-difluorobenzyl) borate anion, tetrakis(4-fluorobenzyl) borate anion, tetrakis(4-ethoxyphenyl) borate anion, tetrakis(4-methoxyphenyl) borate anion, tetrakis(4,5-dimethoxyphenyl) borate anion, tetrakis(4-butylphenyl) borate anion, tetrakis(t-butylphenyl) borate anion, tetrakis(phenyl) borate anion, tetrakis(biphenyl) borate anion, tetrakis(terphenyl) borate anion, tetrakis(mesityl) borate anion, tetrakis(pentamethylphenyl) borate anion, tetrakis(3,5-dimethylphenyl) borate anion, tetrakis(cyclopropyl) borate anion, tetrakis(cyclobutyl) borate anion, tetrakis(cyclohexyl) borate anion, tetrakis(cyclopentyl) borate anion, tetrakis(cyclooctyl) borate anion and tetrakis(phenoxybutyl) borate anion.

7. A polymerization initiator for cationically polymerizable organic substance, characterizing that said polymerization initiator comprises said crystalline ion-association substance claimed in anyone of the claims

1-6.

- 8. The polymerization initiator claimed in the claim 7, wherein said cationically polymerizable organic substance is a compound or mixture of at least two compounds selected from among methylol compounds, ethylenic compounds, polyacetal compounds, organosiloxane compounds, polyamide compounds and heterocyclic compounds.
- 9. The polymerization initiator claimed in the claim 8, wherein said cationically polymerizable organic substance is selected from among organosiloxane compounds, epoxy compounds and mixtures thereof.
- 10. A method of producing the crystalline ion-association substance claimed in anyone of the claims 1-6, characterized in that a metallocene derivative of either mono-nucleus or di-nucleus structure having a general formula (II):

 $\{C_5(R^1)_n\}_{2m}M_m$

wherein, M, C_5 , R^1 , \underline{m} , and \underline{n} have the same meaning as mentioned above, is reacted with a tetradentate borate complex compound having a general formula (III):

 $\{B(R^2)_4\}X$

wherein R^2 has the same meaning as mentioned above and X is an alkali metal atom.